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DETERMINATION OF RADON CONCENTRATIONS IN SOME BUILDING MATERIALS USING PASSIVE TECHNIQUE

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ABSTRACT

Radon is the most important natural radioactive factor harmfully influencing the human population, because radon is radioactive gas comes from the natural decay of uranium deposits in soil, rocks, and water, which is harmful on human and environment. The radon concentrations and exhalation rate were measured using passive technique with CR-39 detector in the building materials from the local market of Dakahlia Governorate. The average values of radon concentrations are ranged from 47.88 to 820.51 Bqm^{-3}. From the obtained results we can conclude that the values of granite higher than the different other materials. This study can be used as reference information to assess any changes in the radioactive background level in our houses and detect any harmful radiation that would affect the human.

KEYWORDS: Radioactivity, Radon, Building Materials, CR-39, Radiation

INTRODUCTION

Natural occurring radionuclides enter the human body mainly by inhalation of radon and thoron gases and by ingestion of primordial radionuclides and their progeny [1]. ^{222}Rn is produced through α-decay of ^{226}Ra in the soil is the only gaseous decay in this series as a noble gas, part of the ^{222}Rn emanates from the soil grains into the air and diffuse to the atmosphere. Construction materials are sources of indoor airborne radioactivity and external radiation from the decay series of uranium in buildings. Radon is perhaps the most important natural radioactive factor harmfully influencing the human population, because radon is radioactive gas comes from the natural decay of uranium deposits in soil, rocks, and water, which is harmful on human and environment [2]. Building and industrial materials, which are brought from the deserts like sands, also contribute to environmental radioactivity in two ways, first, by gamma radiation mainly ^{226}Ra, ^{232}Th, ^{40}K and their progenies to a whole body dose and in some cases by beta radiation to a skin dose, and secondly by releasing the noble gas radon, its radioactive daughters, which are deposited in the human respiratory tract [3]. Radionuclides present in building materials are responsible for both external and internal exposure. The external exposure is caused by gamma rays emitted mainly by radionuclides of uranium ^{238}U and thorium ^{232}Th decay series as well as potassium ^{40}K. The internal exposure is caused by ^{222}Rn and its short-lived decay products. Radon is an alpha emitter that may be easily inhaled and its descendants may be deposited in tissues of the respiratory tract [4]. Radiation include external sources, such as cosmic rays and radioactive materials in the ground, building materials, internal sources resulting from inhalation and ingestion of naturally occurring radioactive materials (NORMs) in air. Radioactive materials are found naturally as trace elements in soil, rocks, building materials, ground water, air and vegetation [5-6]. Most of the radon in indoor air comes from soil underneath the home. As uranium breaks down, radon gas forms and seeps into the house. Radon from soil can get into any type of building homes, offices, schools and build up to high levels in the air inside the

building [7]. ^{222}Rn concentration indoors depends primarily on the construction of the building and the amount of ^{222}Rn in the underlying soil. Daughter nuclides following radon decay, attached to microscopic dust's particles are inhaled and emit alpha particles, which effectively cause biological damages to the lung cells. The continuous damage produced by alpha particles emitted from radon in lungs may cause cancer [8-9].

Radon in indoor spaces may originate from exhalation from rocks and soils around the building or from construction materials used in walls and floors. Radon levels are generally highest in cellars and basements because these areas are nearest to the source and are usually poorly ventilated. Radon can escape out of the ground and build up in confined spaces, particularly underground in basements of buildings, caves, mines and ground floor buildings [10]. Passive technique, which used in the present study based on the registration of alpha tracks from ^{222}Rn on alpha sensitive track detector that was developed for radon exploration. The detector is exposed to the soil gas for a specific period of time. The alpha tracks are registered on the detector and the tracks density gives a measure of ^{222}Rn concentration in the building materials. As it is a very simple technique, it can be implemented easily for field studies, since they do not require electronic system.

The present work is aiming to determine the radon concentrations and radon exhalation rate in the samples of building materials from local market of Dakahlia Governorate, in order to detect any harmful radiation that would affect the human and radioactivity background levels in Egyptian building materials which, can be used as reference information to assess any changes in the radioactive background level in our houses.

MATERIALS AND METHODS

Forty samples from different type of building materials were collected from the local market of Dakahlia Governorate. The samples were measured using passive technique (Can Technique) to determine the values of radon concentration and exhalation rate with CR-39 detector. The rock samples are crushed to a grain size 1 mm, and placed into sample containers. All samples were dried in oven at 110°C for 3hr, minced, sieved by 1-mm mesh, weighted and carefully sealed for 61 days in cylindrical containers made from a good kind of plastic with dimensions of 9 cm in diameter and 16 cm in depth. Each sample container was capped tightly to an inverted cylindrical plastic cover as shown in **Figure 1**. The calculations relied on establishment of secular equilibrium in the samples due to the much smaller life time of daughter radionuclides in the decay of ^{232}Th and ^{238}U series.

A piece of CR-39 of 700 µm thickness (American Technical Plastic, Inc.) detector of area 1.5 cm x1.5 cm fixed at the bottom center of the inverted plastic cover. During the exposure time of α-particles from the decay of radon and their daughters, bombard the CR-39 detector in the air volume of the cylindrical containers. After the irradiation period, the bombarded detectors were collected and chemically etched in NaOH solution 6.25N at 70°C for 7 hr [11].

After etching, CR-39 detectors were washed in distilled water and then dipped for few minutes in a 3 % acetic acid solution, washed again with distilled water and finally air-dried. The track density was determined by using optical microscope [12-13] which calibrated before usages. The background of CR-39 track detector was counted by optical microscope and subtracted from the count of all detectors [14]**.** The value of radon concentration in (Bqm^{-3}) at secular equilibrium is given by the following equation:

$$C_{Rn} = \frac{\rho}{\eta T} \qquad (1)$$

Where, C_{Rn} is radon concentration (Bqm^{-3}), ρ is the track density (track cm^{-2}), T is the exposure time (day), and η is the calibration coefficient of CR-39 nuclear track detectors obtained from the experimental calibration 0.22 ± 0.04 tracks $cm^{-2}day^{-1}/Bqm^{-3}$ of radon, respectively [15-16-17]. The relation gives radon exhalation rate**:**

$$E_A = \frac{CV\lambda}{A(1-e^{-\lambda t})} \quad (2)$$

Where, E_A is the surface exhalation rate in ($Bqm^{-2}h^{-1}$), C is the radon concentration in (Bqm^{-3}), λ is the decay constant of radon (h^{-1}), V is the effective volume of the cup (m^3), A is the area covered by the can (m^2) and t is the irradiation time [18].

$$E_M = \frac{CV\lambda}{M(1-e^{-\lambda t})} \quad (3)$$

Where, E_M is the mass exhalation rate in ($Bqkg^{-1}h^{-1}$), C is the radon concentration in (Bqm^{-3}), λ is the decay constant of radon (h^{-1}), V is the effective volume of the cup (m^3), M is the mass of sample (kg) and t is the irradiation time[19-20-21]. The annual absorbed dose rate (D_{Rn}) was calculated according to the following equation:

$$D\,(mSv\ y^{-1}) = C_{Rn}\,.D.\ H.\ F.\ T \quad (4)$$

Where, C_{Rn} in Bqm^{-3} is the measured mean radon activity concentration in air, F=0.4 is the indoor equilibrium factor between radon and its progeny, H is the indoor occupancy factor (0.4), D is the dose conversion factor ($9x10^{-6}$ $mSvh^{-1}$ per Bqm^{-3}) and T is the indoor exposure time in hours per year which, equal the following value:

$$T = 0.8 \times 24\ h \times 365.25\ days \cong 7013\ h\ y^{-1}$$

The annual effective dose rate was calculated according to the following equation:

$$H_E\,(mSv\ y^{-1}) = D.\ W_R\ .W_T \quad (5)$$

Where D is annual absorbed dose, W_R is radiation weighting factor for alpha particles which equal 20 and W_T is tissue weighting factor for the lung equal 0.12 [22]. The concentration of the radon progeny in air is very dependent upon ventilation and humidity, so that we calculate the working level (WL). Working level exposure estimates determine if workplaces are compliant with recommended exposure limits and also provide guidance on the appropriate level of personal protection needed if administrative and engineering controls are unable to reduce radon levels to acceptable concentrations. The working levels given using the following equation:

$$WL = \frac{C_{Rn} \times F}{3700} \quad (6)$$

Where, C_{Rn} is radon concentration in (Bqm^{-3}) and F is the equilibrium factor for radon has been taken as 0.4 as suggested by [23].

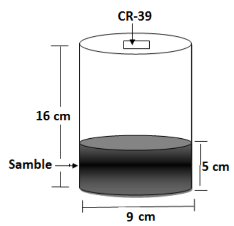

Figure 1: Cylindrical Container Made of Plastic

RESULTS AND DISCUSSIONS

The values of radon concentration (C_{Rn}), area exhalation rate (E_A), mass exhalation rate (E_M) of the building materials are given by **Tables 1**. The values of radon concentrations in Bqm^{-3} are ranged from 123.77 to 266.39 for black cement, 41.95 to 53.80 for white cement, 52.16 to 75.85 for gypsum, 178.68 to 349.77 for sand, 43.95 to 108.71 for ceramic, 150.67 to 271.23 for marble, 65.64 to 152.83 for brick, 359.98 to 366.46 for stone, 47.39 to 113.04 for gravel and 531.44 to 1348.73 Bqm^{-3} for granite.

The values of radon concentrations in Al–Mesalh black cement has a high value but El-Askeria black cement has low value. In the case of white cement the value of radon concentration in Helwan white cement has high value but Sinai white cement has low value. Super Sinai gypsum has high value of radon concentration but Al-Bulah gypsum has low. Yellow sand (Kom Hamada) has a high value of radon concentration but White sand (Al-Sharqia) has a low value. Al-Amear ceramic has a high value of radon concentration but Alpha ceramic has a low value. Green marble (Indian) has a low value of radon concentration but White marble (Turkish) has a high value. The values of radon concentration in Red brick have a high value but Jerry brick has a low value. Hashemi stone (Egyptian) has a high value of radon concentration but Mica stone (Egyptian) has a low value. Al-Sharqei gravel (Salehia) has a low value of radon concentration but Kom Hamada gravel has a high value. Rose granite has a low value of radon concentration but Jandola granite has a high value.

Figure 2 gives the relation between the type of the sample and radon concentration.

The values of area exhalation rate in ($Bqm^{-2}h^{-1}$) and mass exhalation rate in ($Bqkg^{-1}h^{-1}$) of radon were calculated and the average values were given in **Table 2**. The average values of area exhalation rate in ($mBqm^{-2}h^{-1}$) of black cement is 239.42 and 61.84 of white cement,83.74 of gypsum, 331.14 of sand, 107.44 of ceramic, 270.23 of marble, 140.43 of bricks, 467.68 of stone, 113.69 of gravel and 1059.92 of granite.

Figure 3 shows the relation between the sample and area exhalation rate of radon, from the figure we find that the granite sample has a high value. The variation in the values of radon concentrations due to the difference in the chemical composition and the geological form of the samples.

The average values of mass exhalation rate in ($mBqkg^{-1}h^{-1}$) of radon for black cement is 3.22 and 0.86 of white cement, 1.63 of gypsum, 3.12 of sand, 1.43 of ceramic, 3.09 of marble, 1.68 of bricks, 5.46 of stone, 1.17 of gravel and 11.20 of granite.

Figure 4 shows a linear correlation between radon concentration and area exhalation rate. The correlation coefficient is equal R^2 =1, which is a very good correlation. The values of annual absorbed dose and the working level were calculated. The values of working level (WL) and annual absorbed dose (D) of the building materials are given by **Table 1**.

The average values of working levels in (mWL) are ranged from 5.13 to 88.67 and the average values of annual absorbed dose in ($mSvy^{-1}$) of black cement is 2.34 and 0.60 of white cement, 0.81 of gypsum, 3.23 of sand, 1.04 of ceramic, 2.64 of marble, 1.37 of bricks, 4.57 of stone, 1.11 of gravel and 10.34 of granite.

A granite sample has a high value of annual absorbed dose, but White cement has a low value. **Figure 5** gives the comparison between the values of annual absorbed dose of the samples of building materials, from this the annual absorbed dose of Granite has a high value but White cement has a low value.

Table 1: Radon Concentration (C_{rn}), Area Exhalation Rate (E_A) and Mass Exhalation Rate (E_M) for Building Materials

Sample	No.	Sample Name	C_{Rn} (Bq m^{-3})	E_A (mBqm^{-2}h^{-1})	E_m (mBqKg^{-1}h^{-1})	mWL	D (mSvy^{-1})
Black Cement	1	AL-Altamier	189.41 ± 10.05	243.50 ±12.10	3.10 ± 0.16	20.40	2.38
	2	EL- Askeria	123.77 ± 8.19	159.76 ±10.09	2.35 ± 0.15	13.41	1.56
	3	AL-Amerya	147.46 ± 8.94	189.39 ±11.11	2.84 ± 0.17	15.85	1.85
	4	AL-Shora	180.77 ± 9.83	233.19 ±11.09	3.15 ± 0.15	19.57	2.28
	5	AL-Mesalh	266.39 ± 11.99	342.71 ± 14.05	4.23 ± 0.17	28.74	3.35
	6	AL- Waha	207.74 ± 10.55	267.98 ± 12.21	3.79 ± 0.18	22.42	2.62
White Cem.	1	Sinai	41.95 ± 5.36	54.11 ± 6.43	0.81 ± 0.16	4.51	0.52
	2	Royal	47.91 ± 4.76	61.84 ± 6.24	0.78 ± 0.06	5.11	0.60
	3	Helwan	53.80 ± 5.06	69.57 ± 6.76	0.98 ± 0.09	5.82	0.68
Gypsum	1	AL-Medena	62.44 ± 5.81	79.88 ± 6.80	1.49 ± 0.16	6.70	0.78
	2	Sinai	69.97 ± 6.11	90.18 ±7.32	1.68 ± 0.15	7.57	0.88
	3	Al-Bulah	52.16 ± 5.29	66.99 ± 6.12	1.52 ± 0.14	5.66	0.65
	4	Super Sinai	75.85 ± 6.40	97.91 ±7.33	1.84 ± 0.11	8.21	0.95
Sand	1	Yellow (AL-Sharqia)	232.48 ± 11.17	298.90 ±14.11	2.61 ± 0.10	25.08	2.92
	2	White (AL-Sharqia)	178.68 ± 9.83	230.62 ± 11.01	2.32 ± 0.13	19.35	2.25
	3	Yellow (Kom Hamada)	349.77 ± 13.71	450.93 ±16.76	4.27 ± 0.16	37.80	4.41
	4	White (Kom Hamada)	267.43± 11.99	344.11 ± 14.12	3.28 ± 0.12	28.85	3.36
Ceramic	1	Kilopatra	95.30 ± 7.15	122.39 ± 9.77	1.66 ± 0.17	10.22	1.19
	2	AL- Amear	108.71 ± 7.67	140.43 ± 9.75	2.13 ± 0.15	11.71	1.37
	3	Alpha	43.95 ± 4.84	56.68 ± 5.33	0.63 ± 0.05	4.72	0.55
	4	AL-Jeohara	87.18 ± 6.78	112.09 ± 7.44	1.42 ± 0.94	9.44	1.09
	5	Royal	82.33 ± 6.63	105.64 ± 7.55	1.32 ± 0.09	8.80	1.03
Marble	1	Cecilia (Italian)	235.69 ± 11.25	304.06 ± 14.22	3.49 ± 0.13	25.51	2.97
	2	Amblador (Spain)	180.77 ± 9.83	233.19 ± 11.22	2.69 ± 0.15	19.53	2.28
	3	White (Turkish)	271.23 ± 12.07	349.15 ± 15.13	3.72 ± 0.18	29.24	3.41
	4	Green (Indian)	150.67 ± 9.01	194.54 ± 11.27	2.49 ± 0.18	16.31	1.90
Brick	1	White cement	101.71 ± 7.37	131.41 ± 9.01	1.41 ± 0.09	11.02	1.28
	2	Red	152.83 ± 9.09	197.12 ± 11.07	2.52 ± 0.91	16.51	1.92
	3	Thermal	114.60 ± 7.82	148.16 ± 9.44	1.79 ± 0.71	12.40	1.45
	4	Jerry	65.64 ± 5.96	85.03 ± 6.32	0.99 ± 0.07	7.11	0.83
Stone	1	Hashemi (Egyptian)	366.46 ± 14.00	471.55 ± 18.42	6.01 ± 0.92	39.54	4.61
	2	Maica (Egyptian)	359.98 ± 13.93	463.82 ± 16.38	4.93 ± 0.51	38.93	4.54
Gravel	1	AL-Sharqei (Ataka)	112.44 ± 7.74	144.29 ± 9.73	1.83 ± 0.08	12.11	1.41
	2	AL-Sharqei (Salehia)	47.39 ± 5.06	60.55 ±.446	0.68 ± 0.07	5.08	0.59
	3	AL-Sharqei(Abu Hammad)	80.70 ± 6.55	104.35 ± 7.77	1.13 ± 0.08	8.71	1.02
	4	Kom Hamada	113.04 ± 7.82	145.58 ± 9.90	1.57 ± 0.09	12.22	1.42
Granite	1	Rose	531.44 ± 16.84	686.32 ± 20.65	6.94 ± 0.92	57.40	6.69
	2	Jandola	1348.73 ± 26.90	1744.12 ± 33.89	19.98 ± 0.83	145.83	17.01
	3	Ghazal	589.79 ± 17.80	759.11 ± 21.33	7.68 ± 0.92	63.67	7.42
	4	Maltk Brown	812.09 ± 20.86	1050.14 ± 25.53	10.95 ± 0.82	87.78	10.24

Table 2: The Average Values of Radon Concentration (C_{rn}), Area Exhalation Rate (E_A), Mass Exhalation Rate (E_M), Working Level (WL) and Annual Absorbed Dose (D) for Building Materials

Sample	C_{Rn} (Bq m^{-3})	E_A (mBq m^{-2} h^{-1})	E_M (mBq Kg^{-1} h^{-1})	mWL	D (mSvy $^{-1}$)
Black Cement	185.92	239.42	3.22	20.03	2.34
White Cement	47.88	61.84	0.86	5.13	0.60
Gypsum	65.10	83.74	1.63	7.00	0.81
Sand	257.09	331.14	3.12	27.75	3.23
Ceramic	83.49	107.44	1.43	8.96	1.04
Marble	219.59	270.23	3.09	22.62	2.64
Bricks	108.69	140.43	1.68	11.75	1.37
Stone	363.22	467.68	5.46	39.20	4.57
Gravel	88.39	113.69	1.17	9.52	1.11
Granit	820.51	1059.92	11.20	88.67	10.34

Table 3: The Comparison between the Obtained Results and the Published Data for Building Materials in Different Countries

Country/ Org.	Sample Name	C_{rn} (Bqm^{-3})	E_x (Bqm^{-2}h^{-1})	D (Msvy^{-1})	References
Egypt	Marble Granite		0.33 -1.25 0.29 -1.10		[23]
Egypt	Black Cem. White Cem. Sand Ceramic Bricks Gypsum	144 136 108 192 137 71	0.093 0.088 0.070 0.124 0.088 0.046	0.24 0.11 0.17 0.62 0.35 0.10	[24]
Iraq	Stone	121	0.72		[19]
Iraq	Sand Gravel Gypsum	480.71 391.69 284.86	0.345 0.272 0.156		[25]
Iraq	Marble	426	0.324		[26]
Palestine	Marble Ceramic Graval Granit Gypsum	240 193 69 322 31	0.438 0.347 0.126 0.589 0.055	6.06 4.88 1.75 8.12 0.79	[27]
India	Black Cem. White Cem.	332 550	0.263 0.299	0.53 0.62	[28]
India	Bricks Granit Sand	190 300 238			[29]
K. S. A	Granite		0.33 -1.25		[30]
Greek	Granite		1.24 -3.54		[31]
ICRP		200 - 600			[32]
Egypt	Black Cem. White Cem. Sand Marble Ceramic Graval Bricks Granit Gypsum Stone	186 48 257 210 83 88 109 820 65 363	0.238 0.061 0.331 0.270 0.107 0.113 0.140 1.059 0.083 0.467	2.34 0.60 0.81 3.23 1.04 2.64 1.37 4.57 1.11 10.34	The present study

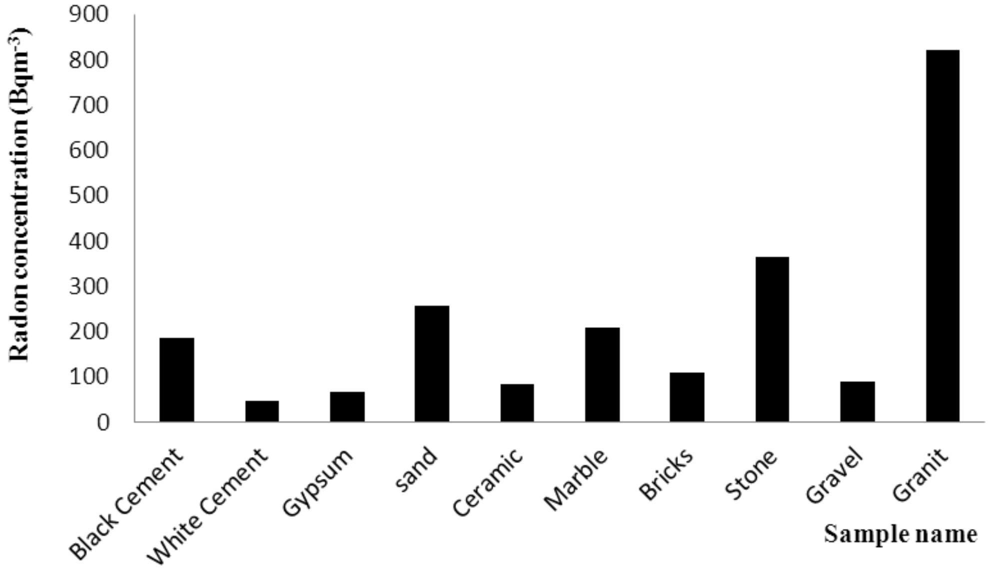

Figure 2: The Relation between the Sample Name and Radon Concentration of the Building Materials

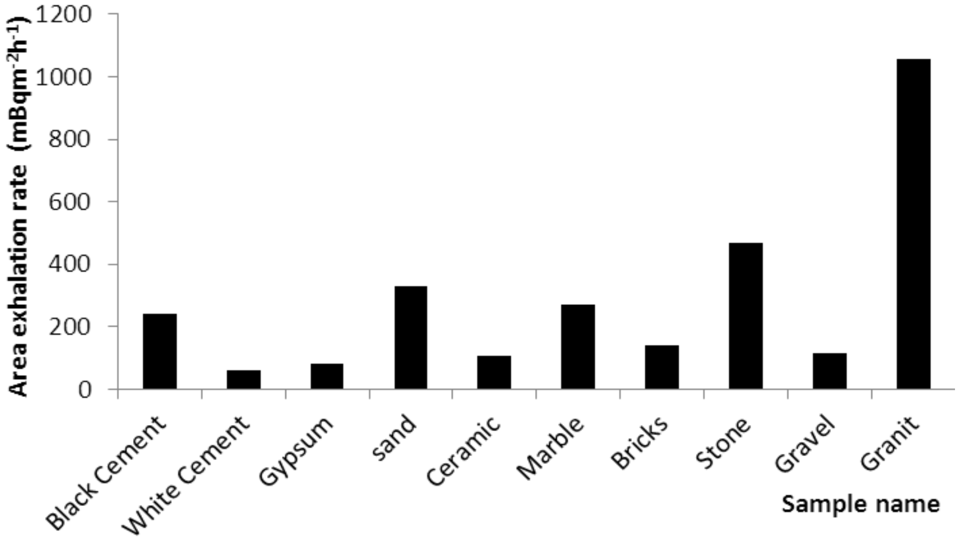

Figure 3: The Relation between the Sample Name and Area Exhalation Rate of the Building Materials

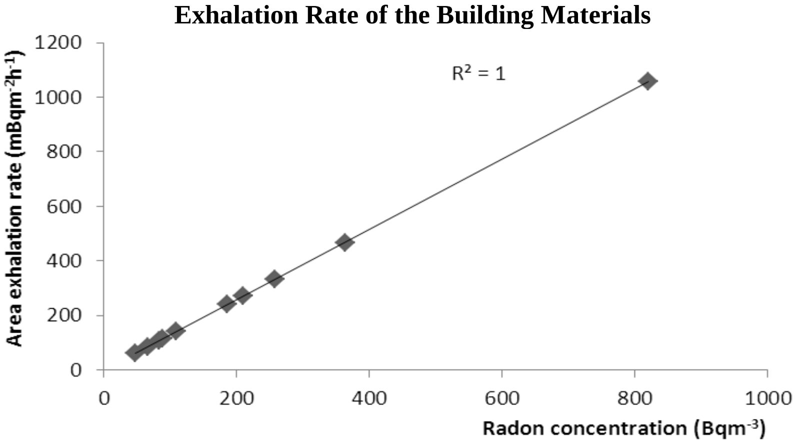

Figure 4: The Correlation between Radon Concentration and Area Exhalation Rate of the Building Materials

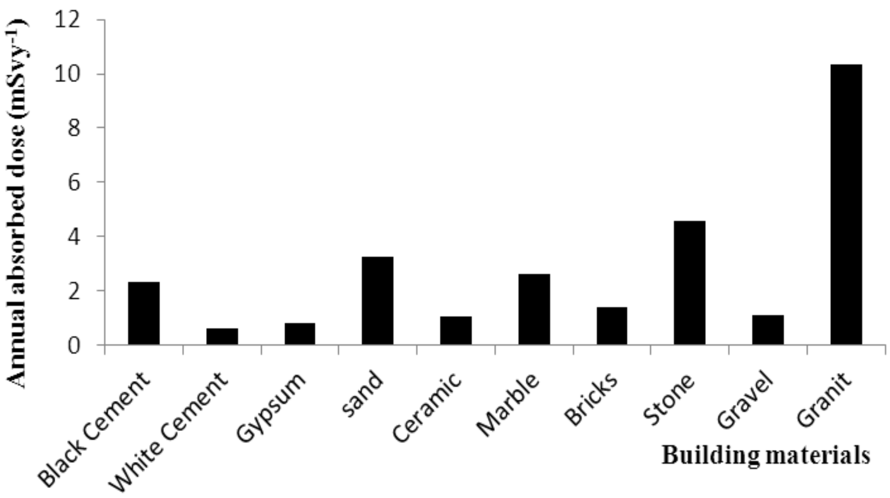

Figure 5: The Comparsion between the Annual Absoebed Dose of the Building Materials

CONCLUSIONS

This study can be used as reference information to assess any changes in the radioactive background level in our houses and detect any harmful radiation that would affect the human. From the obtained results we find that granite samples have high concentration of radon than the other building materials. The variations in the values of radon concentrations due to the difference in the chemical composition and the geological form of the samples.

The measurements of radon exhalation rate are good indicator for the radon concentrations present in the samples. The obtained results showed that the radon exhalation rate varies linearly with radon concentration as shown in **Figures 4.** Most of the indoor radon values lie in the range of action levels from 200 to 600 Bqm^{-3} recommended by the International Commission on Radiological Protection [35]. But in the granite samples are higher than the recommended limit.

The upper limits for indoor radon concentration in most European countries are 200 Bq m^{-3} for new buildings and 400 Bq m^{-3} for old buildings [36]. This value is below the radon reference level which ranges from 200–600 Bq/m^{3} as recommended by ICRP, IAEA, and is lower than the USA intervention radon level.

The values of annual absorbed dose higher than the reported worldwide values which indicate the safe use of these sediments as building materials which, equal 0.3 mSvy^{-1} [7]. The corresponding worldwide average value is equal 1mSvy^{-1} [4] and corresponds to the annual effective dose equivalent of 1 mSvy^{-1} for general public [37].

The dose limit of the permissible effective dose from occupational radiation exposure is 20 mS y^{-1} in all European countries (in the USA 50 mS y^{-1}). From the previous discussion we conclude that the results are agreement with the published data for building materials in different countries as shown in **Table 3**. In the end we can protect ourselves from radon risk and harmful effects on the human body by ventilation of our house.

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